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An assessment of options for CO_2 removal from the atmosphere

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Abstract

Recent work examining likely changes in global temperatures as a result of carbon dioxide (CO_2) emissions has suggested that cumulative CO_2 emissions (i.e. total emissions over an extended period of time) could be more significant than the differences between particular emissions pathway (e.g. with different timing of emissions or peak emissions rate) in determining how the global climate might change in response to CO_2 emissions. This suggests that effective measures to mitigate the risk of dangerous climate change will need to limit cumulative emissions of CO_2. Further, if cumulative CO_2 emissions overshoot acceptable limits, it will become necessary to remove CO_2 from the air – so-called 'negative emissions'. Technologies that effect 'negative emissions' could also be used to offset additional anthropogenic emissions from sectors where greenhouse gas emissions are difficult or impossible to reduce beyond certain, still relatively high, limits. If the prevailing carbon price for marginal abatement options rises significantly from current levels (e.g. of order up to \$200/tCO_2 has been suggested by some) then a relatively wide range of options for removing CO_2 from the air may become cost-effective. Additionally, some options for removing CO_2 from the air are likely to have much lower abatement costs. This paper summarises results from research conducted to compare and contrast various options for capturing CO_2 from the air, with a particular focus on establishing the potential of these options to have a significant impact in reducing CO_2 emissions and, if so, over what timescales.

Keywords: towards negative CO_2 emissions; biomass with CCS; captuting CO_2 from the air; mineralisation

1. Introduction

CO_2 is long-lived in the atmosphere, and it seems increasingly likely that CO_2 emissions will overshoot the limit on the cumulative total that is likely to be needed to limit a global temperature rise to below 2°C above pre-industrial levels [1]. It may, therefore, become necessary to remove CO_2 from the atmosphere. Additionally, it has been suggested that emissions of greenhouse gases in developed countries may need to be cut by at least 80% by 2050 to mitigate the risk of dangerous climate change [e.g. 2]. It will be difficult or impossible to achieve such a significant reduction in direct emissions in some sectors (e.g. agriculture and food production, cement production and air and/or marine transport) and this again suggests a role for 'negative emissions' options that can be used to offset emissions generated by activities that have no cost-effective abatement option available. A broad range of options have been identified as having potential to remove CO_2 from the atmosphere and are at various stages of development.

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This paper uses case studies to provide a cross-section of these alternative strategies for achieving negative emissions. They are:

- Artificial trees;
- Soda/Lime process;
- Augmented ocean disposal process;
- Biochar; and
- Biomass Energy with Carbon Capture and Storage (BECCS).

It should be noted that the work presented here is primarily based on a literature survey and there remain key uncertainties/gaps. Considerable further work is required in certain areas. The conclusions should therefore be regarded as preliminary and subject to revision in the light of further research.

2. Definitions and classes of technologies for capturing CO_2 from the air

Air capture of CO_2 involves the separation of a gas at low partial pressure, from a mixture of inert and not so inert gases. Separation of gases, in general, can be conducted using one of only a few methodologies. These include membrane separation, selective condensation, and chemical scrubbing. Of these, due to the low ambient partial pressure of CO_2 in the atmosphere, the first two methods can be ruled out immediately; in both cases there would be a requirement to pressurise large amounts of the atmosphere, which is it is reasonable to assume would be uneconomic. However, CO_2 is a sour gas and therefore reacts readily with bases of all kinds. As a result, many alkali scrubbing agents are able to reduce the residual partial pressure of CO_2 in a gas stream to very low levels. Chemical scrubbing is a viable air capture methodology and, directly or indirectly, all proposed air capture methods rely on chemical scrubbing of some kind. This may be using simple scrubbing agents such as sodium hydroxide (NaOH) in a scrubbing tower; or calcium hydroxide [$Ca(OH)_2$] dissolved in sea water. The linkage is more tenuous with biomass based systems, but photosynthesis can be seen as natural chemical scrubbing.

Chalmers and Gibbins [3] categorised carbon capture systems into three classes: 1, 2 and 3 – depending on whether they are: carbon positive (despite the addition of some CCS); near carbon neutral; or potentially carbon negative (air capture), respectively. Class 3 was further subdivided into classes 3A and 3B. These are:

- 3A systems are those that capture CO_2 directly from the air;
- 3B systems are those that use biomass in a conventional CCS power plant of some kind – i.e. Biomass energy + CCS (BECCS).

All forms of air capture are fundamentally post-combustion scrubbing techniques. A critical issue is whether or not the captured CO_2 is sequestered in geological formations. This is an important distinction, as to sequester fluid CO_2 into an underground sink the CO_2 must be compressed to high pressure. A significant proportion of the net work input into the process is consumed in this final stage, and processes that do not need to compress CO_2 could, at least in principle, be cheaper to operate. Further, the investment in compression and injection equipment can be avoided, and a reduced CO_2 transportation network has the potential to lead to further cost savings. As a result, class 3A has been split into two distinct divisions (i) 3AA systems where CO_2 is compressed for geological storage); and (ii) 3AB systems where CO_2 at low partial pressure is fixed in a stable, form and then stored geologically).

The examples of 3A technologies selected for further study have been chosen to represent the major types of air capture. The choice is not intended to be an endorsement of a particular approach or for that matter of the principal architects of the technology. However, when selecting the methodologies, those areas and techniques supported by peer reviewed articles were favoured. BECCS is more developed as a concept and, therefore, the examination of 3B systems in this paper is limited to this generic concept.

3. Class 3AA: Artifical trees and soda/lime process

Class 3AA technologies produce a stream of essentially pure CO_2 at pressures potentially suitable for geological storage. These methods can be viewed as a subset of traditional post-combustion CCS technology as they all use some form of scrubbing system. The apparatus performing the scrubbing can be either static, relying principally on wind to effect mass transport of air across an absorbent; or use a more traditional scrubbing/spray tower where air is entrained by a falling liquid/slurry absorbent. A key cost of these technologies is that the CO_2 collected in the absorbent must be desorbed, compressed, transported, and injected into a geological sink. An example of each of these methodologies is examined in this Section: artificial trees (Section 3.1) and a soda/lime process (Section 3.2).

3.1. Artificial trees

An artificial tree is a device which mimics the processes used by biological plant life to remove CO_2 from the atmosphere. In nature, plants combine CO_2 from the atmosphere with water from their sap chemically, forming various hydro and oxy-hydrocarbons. These chemicals are used as the 'building blocks' of the fibrous or woody stems of the plant. However, in the case of artificial trees, the output from the 'tree' is a stream of essentially pure CO_2 at high pressure, ready for sequestration in the 'normal' manner. Energy is required to effect the chemical transformation in both real and artificial trees – photosynthesis for the former. An industrial energy source is, therefore, required to drive artificial trees and this represents a major cost of the technology. Energy is also required to compress the CO_2 collected in the trees to high pressure so that it can be transported to a site suitable for geological storage. A number of suggested configurations have been proposed, but in all cases the basic methodology is the same.

A significant contributor to developing the concept of artificial trees has been Lackner [e.g. 4]. Lackner's trees are essentially passive devices that present to the atmosphere a large surface area of CO_2 absorbing materials – akin to the leaves of natural trees. Wind is used to drive a current of CO_2 laden air across the trees' absorbing surface, so that mass transfer of CO_2 to the absorbent takes place. The current of wind then carries the CO_2 denuded air from the trees' surface. As a result, the wind provides sufficient air change to expose the absorbent to fresh, and hence CO_2 'rich', air continually. The absorbent is supported on a substrate and is recycled as it becomes saturated and regenerated either thermally or using moisture swing in the base of the tree – releasing essentially pure CO_2. This CO_2 can then be compressed and transported to sites suitable for geological storage, probably using pipelines given the likely distributed nature of the trees since many trees would be required to effect an environmentally significant amount of carbon capture.

The trees can be arranged as a series of discrete, sail-like structures. Practically any geographic location is feasible, provided there is a source of energy to power the trees, water if found to be necessary and also reasonable access to a suitable site for CO_2 storage. One suggestion, is to use wind turbines, built adjacent to the trees to provide the electrical power for the absorbent regeneration process particularly in areas when wind resource is 'stranded' due to difficulties in identifying an commercial option for exporting power from that area. This configuration could lead to savings in planning and installation cost, with artificial trees able to take power as it becomes available from the wind turbine. The trees might then become part of a load matching system helping smooth out the variable supply curve of wind power systems. However, if the trees themselves are utilised at less than 100% duty cycle there would be an inevitable increase in their notional capital cost per unit of capture. Of course, it would also still be necessary to maintain a connection with a CO_2 transport network.

3.2. Lime/soda process

The lime/soda process developed by Keith and co-workers [e.g. 5] technology is a direct chemical scrubbing techniques that can also be viewed as a cousins of conventional post-combustion capture systems that are being explored for CO_2 capture at power plants. As illustrated in Figure 1, the technique is a cyclic process that requires energy input alone (fuel rather than electricity) and outputs a stream of essentially pure CO_2 that can then be compressed and exported into a CO_2 transport network. The technology components are well understood and exist in other industries. The technology could, however, be quite capital-intensive given the dilute nature of the feedstock and the need for a high temperature calciner.

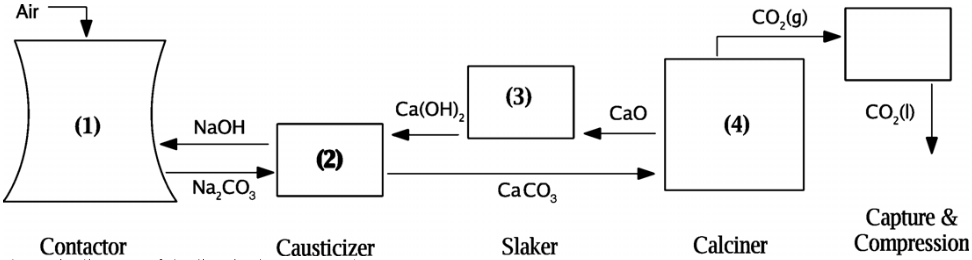

Figure 1 Schematic diagram of the lime/soda process [5]

The underlying chemistry of the lime/soda process consists of four discrete reactions. The first step is the absorption of CO_2 from air using sodium hydroxide (NaOH) generating soda (Na_2CO_3) in a scrubbing tower or contactor. The soda solution generated in the tower is then reacted with calcium hydroxide [$Ca(OH)_2$] in a reactor called a causticizer, to regenerate the sodium hydroxide which is returned to the contactor. This is the lime-soda reaction. The waste product from this process is calcium carbonate ($CaCO_3$), which precipitates and is then filtered from the sodium hydroxide solution continually. This calcium carbonate is then converted to lime (CaO) using the calcination reaction, slaked and returned to the causticizer. These processes are repeated indefinitely. The output from the process is a stream of CO_2 generated in the calciner, which if fossil fuel fired must have an associated CCS system of some kind to maximise the negative emissions of the overall system.

Although the lime/soda process has some similarities with the use of artificial trees, there are also some important differences. For example, Keith et al [5] propose a more traditional contactor for the primary absorption stage. This means that the primary materials needed (concrete and steel) are readily available, and no special construction processes are required. Additionally, the CO_2 saturated absorbent is regenerated using a two stage chemical process, followed by calcination requiring substantial heat addition. Two chemical loops are embodied in the process and it is claimed this offers thermodynamic advantages as each process can be operated close to equilibrium.

4. Class 3AB: Augmented ocean disposal process and biochar

Class 3AB technologies sequester CO_2 at low partial pressure by fixing the CO_2 in a stable mineral form, which is then stored geologically. These processes may and often do involve traditional CCS at least in part, as they generally require fossil fuel to supply the necessary energy input. As a result, a traditional CCS plant is required to collect CO_2 if the energy input is sourced from fossils fuels. However, the amount of fossil CO_2 generated is generally less than the amount of CO_2 collected from the atmosphere. An advantage of these processes is that the CO_2 caught from the atmosphere does not require compression to high pressure, and therefore, there is the potential for a net saving in energy terms. The costs associated with transportation and injection might also be reduced reduced. Specific technologies that fall into this class and are considered as illustrative examples in this Section are an augmented ocean disposal process (Section 4.1) and biochar (Section 4.2).

4.1. Augmented ocean disposal process

CO_2 is absorbed by the oceans naturally, but this process can be accelerated by artificially increasing the pH of surface waters with the addition of alkali. This can be achieved using lime, or a similar alkali, which would be manufactured using techniques already practiced by the cement industry and then the dispersal of that lime in areas of the ocean with significant circulation. There is, therefore, an interesting opportunity for process integration using spent lime sorbent from solid looping CCS processes as the feedstock for this process. Since this integrated approach builds upon a system with a primary product (power and possibly heat) there could be potential for this option to find earlier deployment than other approaches to removing CO_2 from the air that do not produce a saleable product.

When alkali is added to seawater, it is expected that the capacity of the oceans to absorb CO_2 will be increased. In fact, a range of weathering techniques which aim to chemically enhance the ability of oceans to absorb CO_2 have been proposed. For example, Kruger [6] summarises a variety of these techniques such as HCl removal from the oceans, enhanced carbonate dissolution, accelerated weathering of silicate minerals and the "Cquestrate" process. Fertilisation to enhance ocean biological productivity has also been considered.

Augmented ocean disposal processes are promising, but further work is required to improve understanding of how their implementation would affect the marine environment. Some important factors determining both the potential magnitude of their impact and also rate of deployment are the ability to extract sufficient limestone/dolomite for lime production and identifying and/or developing infrastructure for transport of fuel and mineral to/from the calcinations plant, almost certainly including appropriate ships and deep water ports. While neither of these considerations are 'show-stoppers' they do indicate that significant upfront investment would be required as part of any widespread deployment of augmented ocean disposal processes.

4.2. Biochar

Biochar is produced by the combustion of biomass in a low-zero oxygen environment, called pyrolysis. This produces three products: a solid fraction known as char; a liquid fraction; and a gaseous fraction. The latter two products can be used to generate energy and the char can be land filled or used to enrich agricultural land, where it has been known to enhance crop yields. Biochar could allow CO_2 to be removed from the air if carbon is effectively locked in to the soil. An attraction of biochar is that its multiproduct nature provides a driving force for early deployment. There is, however, some uncertainty on how much char would be used as a soil conditioner and how much might find its way into BECCS-type applications.

The process technology involved in producing biochar is considered to be small scale and non-capital intensive, so the process lends itself to farmers, small landowners and local authorities in developed nations and could assist in rural diversification and poverty alleviation in developing nations [7]. Of course, the benefits of small scale technology must also be weighed up against the increased number of individual installations that are then required for a similar level of CO_2 emissions reduction to be achieved. The scalability of biochar as a CO_2 mitigation option will depend on a number of factors including available biomass feedstock, scability of pyrolysis process technology and logistical considerations for large scale biomass feedstock supply chains.

There are also substantial uncertainties with the pyrolysis process technologies. Present projections are based on small scale systems and the impact of scaling up processes, improvements from learning by doing and agronomic/yield benefits of adding biochar to soils make the economics of biochar production at the commercial scale difficult to deduce. Additionally, further work is required to improve understanding of the influence of pyrolysis process conditions and feedstock on biochar yield and stability; the stability of carbon in the char and its interactions in different conditions; and storage capacity of the char in soils and effects on yields in different climates.

5. Class 3B: biomass energy with carbon capture and storage (BECCS)

Class 3B CCS is most likely to be delivered in biomass energy with carbon capture and storage (BECCS) schemes. A typical example is the direct combustion of (low grade) biomass fuels in a conventional power plant with the capture of greenhouse gas (GHG) emissions generated using carbon capture and storage (CCS) technology that is also being developed for use at power plants burning fossil fuels. Of the case study technologies considered in this report BECCS has the greatest technology maturity and could be introduced relatively easily in today's energy system. The presence of a saleable product (e.g. electricity from a biomass fired power plant) also contributes to making this an attractive option for removing CO_2 from the air. It also likely, however, that BECCS will require appropriate policy support and integration with general CCS strategy for significant commercial-scale deployment to occur.

It is, of course, important to note that some significant challenges must also be overcome before widespread, commercial deployment of BECCS occurs. As with biochar processes, further work is required to understand the full lifecycle emissions associated with BECCS projects including feedstock production, harvesting, processing and transport to the power plant (or other facility) where it is combusted. It is likely, however, that monitoring, reporting and verifying CO_2 stored in a class 3B BECCS project will be able to use many of the standard guidelines that can be expected for 'conventional' CCS projects with fossil fuels. This contrasts with biochar, for example, where there is more significant uncertainty over how much carbon it is reasonable to assume has been retained in soil and over what timescales.

One significant concern associated with significant use of BECCS is that there must be a sufficient supply chain to avoid significant conflicts with food supply and other important uses of biomass and the land that may be used to grow it as an energy crop. If carefully managed, the development of a BECCS supply chain could, however, have a positive influence in establishing and sustaining a robust global agricultural sector supporting both food production and biomass supply. This could include improving management of land use with better soil restoration, the creation of vegetation filters and possible reduction of wildfire risk. There is also potential for poverty reduction by stimulating stagnant agricultural sectors especially in equatorial regions [7], but these positive outcomes would require significant reorganisation of the international trade system [8]. The potential use of waste as a BECCS feedstock would also result in probable improvements in waste management and resource efficiency as the economic value of waste products is realised [9].

6. Discussion and conclusions

This paper presents results from a critical review of literature assessing various options for capturing CO_2 from the air on a full-cycle basis. The scope focuses on a range of emerging technologies that capture and store CO_2 originating from the air, including the capture of CO_2 emissions from biomass/biofuel use (including co-utilisation of biomass with fossil fuels) and the direct capture of CO_2 from the air. In particular, five case studies are used to allow a detailed review of technology potential to be undertaken, but with a reasonable range of options for removing CO_2 from the air considered.

Some key considerations in determining the scope for commercial deployment of each of these case study options are summarised in Table 1. Overall, it appears likely that some options for delivering 'negative emissions' have significant technical potential for making a cost-effective contribution to mitigating global CO_2 emissions. In particular, class 3B BECCS projects could be rolled out at commercial-scale at a similar speed to other CCS projects based on other fuels. The range of class 3A routes for achieving direct capture from the air also show significant potential, but further work is typically required before these options could be considered for commercial-scale demonstration and rollout.

Table 1 Summary of case studies considered

Technology	Summary	Comments on Scope for Commercial Deployment
Artificial Trees	Emulate action of natural trees by absorbing CO_2 directly from the atmosphere using chemical absorbents, relying principally on wind to effect mass transport of air across an absorbent. The absorbent is then regenerated, releasing almost pure CO_2	Early stage technology, so further development required before commercial viability can be accurately assessed. Geographically distributed network could be beneficial, but potential for planning problems if not located close to CO_2 storage sites (as with many CCS options).
Soda/Lime process	Direct chemical scrubbing technique using a traditional absorption/scrubbing tower arrangement for the primary absorption stage. The CO_2 saturated absorbent is regenerated using a two stage chemical process, followed by calcination.	Further scale-up required, but currently seems likely to be capital intensive, which may be challenging for investment. As with artificial trees and other methods requiring CO_2 storage, potential for distributed network but need to consider location with respect to CO_2 storage sites.
Augmented ocean disposal process	Accelerate CO_2 naturally absorbed by the ocean by artificially increasing the pH of surface waters with the addition of alkali	Further work required to fully understand likely consequences on the marine environment. Rollout potential also likely to be influenced by the rate at which appropriate ships can be built.
Biochar	Biochar is one of three products produced by the combustion of biomass in a low-zero oxygen environment, which can then be land filled or used to enrich agricultural land, effectively locking-in the carbon.	Potential bioenergy related environmental impacts require further investigation, including full life cycle analysis of biochar production from different feedstock streams. More research is also needed on the stability of carbon in the char and its interactions in different conditions. Biomass availability needs to be assessed to determine potential magnitude of use.
Biomass energy with carbon capture and storage (BECCS)	Direct combustion of (low grade) biomass fuels in a conventional power plant with the capture of CO_2 emissions generated using conventional CCS technology. This option is relatively well developed and can be deployed at commercial-scale today.	Some bioenergy related environmental impacts also require further investigation for BECCS (e.g. due to land use change). CCS technology development is obviously also critical in assessing timing for rollout. Biomass availability needs to be assessed to determine potential magnitude of use.

A suggested priority going forward is more detailed work on the costs of the more forward looking technologies, to include R&D pilot and scale-up support, and proper life cycle analyses. This is essential if these technologies are going to be available in the timescales that may be needed to make a meaningful contribution to limiting cumulative CO_2 emissions to the atmosphere. Additionally, if BECCS is to be considered part of the commercial energy mix in the short to medium (as well as longer) term, appropriate policy support and integration with the general CCS strategy should be deliberated urgently.

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